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In re U.S. Patent Application

Stephen ATKINSON

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Attny Docket No.: ATKI3001/ESS

**For: METHOD FOR THE RECOVERY OF
HYDROCARBONS FROM HYDRATES**

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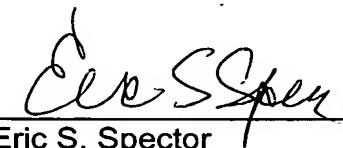
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A copy of the certified copy of priority document UK 0123409.5 filed with WIPO, is enclosed. Please note the received stamp by WIPO of 28 October 2002 for the priority document. This counts for the U.S.A.

Application No. 10/807,152
Attny. Docket No. ATKI3001/ESS

Acknowledgement of receipt of a certified copy of the priority document, is
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Respectfully submitted,
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Case No. P9209US-WO
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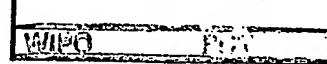
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3. Full name, address and postcode of the or of each applicant (*underline all surnames*)

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The Netherlands.Patents ADP number (*if you know it*)

752764(00)

If the applicant is a corporate body, give the country/state of its incorporation

4. Title of the invention

METHOD FOR THE RECOVERY OF HYDROCARBONS FROM HYDRATES

5. Name of your agent (*if you have one*)

W.H. BECK, GREENER & CO.

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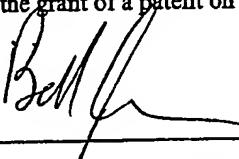
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METHOD FOR THE RECOVERY OF HYDROCARBONS FROM HYDRATES

The present invention relates to a method for recovering hydrocarbons trapped in hydrate formations, and in particular 5 to a method for the recovery of natural gas. It also relates to a fracturing fluid for use in hydrocarbon recovery.

Hydrate formations are an example from a class of chemical compounds known as clathrates. A clathrate is a 10 form of compound in which one component is enclosed within the structure of another. In a hydrate formation, molecules of hydrocarbon (such as natural gas) are trapped in cage-like structures formed by frozen water. Although the water molecules and the hydrocarbon molecules do not interact. 15 chemically, the hydrocarbon molecules are completely surrounded by the water molecules. The structure of the frozen water must therefore be broken down in order to free the hydrocarbon.

Hydrate formations can be found in vast quantities in the earth's crust, principally in seabed sediments and in permafrost regions of the world. Natural gas reservoirs are thought to underlie many of these hydrate formations, the 20 hydrate formations having been formed when rising natural gas met subterranean water deposits, which then froze to enclose 25 the gas.

It is estimated that trillions of cubic metres of natural gas are trapped in subterranean hydrate formations, 30 either sub-sea or on land. Accordingly, there is a huge

potential source of energy if the trapped natural gas can be recovered from these hydrate formations.

Although sub-sea hydrate formations can be located 5 relatively easily by methods such as seismic profiling of the seabed, it is very difficult or at least very expensive to extract the gas from the hydrate formation. Various methods have been developed to address this problem.

10 US 4,007,787 discloses a process for recovering natural gas from gas hydrate reservoirs by passing light hydrocarbons through the formation to dissolve and recover the natural gas. A freezing point depressant such as methanol, ammonia or ethylene glycol can also be injected into the reservoir to 15 accelerate development of production.

Other prior art methods which are disclosed in the introduction to US 4,007,787 include decomposing the hydrate formation by reducing the ambient pressure. The advantage of 20 this method is that the structure of the hydrate can be broken down without having to increase the temperature. However, depressurisation methods are on average about 25% more expensive than conventional gas production methods.

25 Another method of liberating natural gas from hydrate formations is by heating the formation in order to melt the hydrate and free the trapped gas. These "thermal" methods are very expensive however (perhaps twice as expensive as conventional gas production methods) due to the difficulty of 30 heating a formation which is located hundreds of metres under

the permafrost, which may itself be many thousands of metres under sea level.

The third method discussed in US 4,007,787 involves the
5 introduction into the gas hydrate formation of freezing point depressants such as methanol, in order to melt the hydrate and free the natural gas. However, this method is also expensive, due to the cost of the freezing point depressant, and inefficient, since the depressants used to date do not
10 lower the freezing point of the hydrate by a great enough extent to liberate an economically sufficient amount of natural gas.

US 4,424,866 discloses a method of recovering natural
15 gas from gas hydrate formations by pumping a hot supersaturated solution of calcium chloride or calcium bromide under pressure into the hydrate formation so as to hydrostatically fracture the formation, dissolve the solid hydrate and release the gas. However, such "hot solution"
20 methods are rather uneconomical.

In the method disclosed in US 5,713,416, an acidic liquid and a basic liquid are combined and react exothermically to form a hot salt solution, which is injected
25 into a gas hydrate formation in order to decompose the hydrate and release the gas. This method is also uneconomical.

Even if one of the prior art methods is used to extract
30 the gas from the hydrate, further problems can arise in

transporting the liberated gas back to the ship or rig for refinement and storage. The difficulty is that the liberated gas will be wet, i.e. it will carry with it a proportion of water vapour from the hydrate formation, and, at sub-sea 5 temperatures and pressures, the water vapour is likely to freeze, thereby re-forming gas hydrates, which can block the pipeline.

In accordance with the present invention, there is 10 provided a method for recovering hydrocarbon trapped in a hydrate formation, comprising the steps of

(a) contacting the hydrate formation with an aqueous solution of from 10% to 75% by weight of a formate salt of 15 any alkali metal or an acetate salt of any alkali metal, or a mixture of any two or more thereof, whereby said aqueous solution liberates hydrocarbon from the hydrate formation;

(b) transporting liberated hydrocarbon and at least part of 20 said aqueous solution to a separator, wherein during the transportation step said aqueous solution absorbs at least part of any water vapour present to form dilute aqueous solution, thereby inhibiting formation of hydrocarbon hydrates;

25 (c) separating hydrocarbon from said dilute aqueous solution;

(d) regenerating said aqueous solution by heating said 30 dilute aqueous solution to remove absorbed water vapour; and

(e) recycling said aqueous solution to step (a).

For the avoidance of doubt, the phrase "a mixture of any
5 two or more thereof" means a mixture of two or more of any
possible salt, i.e. it encompasses (separately) the following
mixtures: a mixture of different alkali metal formate salts;
a mixture of different alkali metal acetate salts; and a
mixture of formate and acetate salts.

10

For the avoidance of doubt, "from 10% to 75% by weight"
refers to the total amount of salt in said aqueous solution.

Although formate or acetate salts of any of the alkali
15 metals can be used in the method of the present invention, it
is economically advantage to use formate or acetate salts of
potassium, sodium, rubidium or caesium (caesium, although
expensive, is preferred for use in high density drilling
applications). The most preferred salt is potassium formate.

20

The use of aqueous solutions of these particular salts
has a number of advantages. First, the aqueous solutions are
very effective freezing point depressants. This means that
said aqueous solution does not need to be heated in order to
25 decompose the hydrate (although in some circumstances it
might be advantageous to heat the solution). Said aqueous
solution can therefore be injected through the permafrost
into the hydrate formation without melting the permafrost and
thereby risking a gas leak from the well.

30

Secondly, the aqueous solutions of the particular salts themselves have very low freezing points (see Figure 1). This means that said aqueous solution can be used at a very low temperature, again minimising the risk of a gas leak,
5 without risking solidification of said aqueous solution in the well bore. In addition, the aqueous solutions will not freeze in storage tanks when used in the extreme cold such as is found in the Arctic.

10 Thirdly, the aqueous solutions of the said salts have the additional advantage of having vapour absorbent properties. When the vapour which is absorbed is water vapour, said aqueous solution can be thought of as acting as a dehydrating agent. The water vapour which is mixed with
15 the liberated natural gas is therefore absorbed by said aqueous solution, forming a dilute aqueous solution, and thereby drying the natural gas. This has the effect that the re-formation of gas hydrates in the pipelines leading back to the rig is suppressed. Additionally, the absorption of water
20 vapour by said aqueous solution is accompanied by an increase in temperature of said aqueous solution, which itself acts to inhibit formation of gas hydrates.

Lastly, the solutions in question have a very low
25 corrosive tendency and very low environmental toxicity.

Typically, carbon dioxide is present in the liberated natural gas, and this dissolve in said aqueous solution and lower its pH, thereby increasing its corrosive tendency.
30 This problem can be addressed either by admixing an alkali

(such as potassium or sodium hydroxide), preferably to said aqueous solution before it is recycled, to increase the pH to somewhere in the range 8 to 11, or alternatively to add a corrosion inhibitor such as a monohydric alcohol, a 5 polyhydric alcohol, a triazole compound, an alkali metal molybdate, or a mixture of two or more thereof.

Said aqueous solution may be injected into the hydrate formation through an insulating jacket, the jacket having a 10 layer of insulation fluid comprising an aqueous solution of a formate salt of any alkali metal or an acetate salt of any alkali metal, or a mixture of any two or more thereof. This provides an additional means of ensuring that the permafrost does not melt, and means that, if necessary, said aqueous 15 solution can be heated prior to injection into the hydrate formation.

An electrical submersible pump can be placed at the bottom of the well in order to reduce pressure on the hydrate 20 formation, or to stimulate higher levels of gas production.

The arrangement may be such that said aqueous solution is circulated down tubing suspended in a cased well in order to displace said dilute aqueous solution from the hydrate 25 formations, thus reducing the need for pumping either into or out of the well. The natural gas liberated from the hydrate may assist in carrying said dilute aqueous solution from the hydrate and into the pipeline, thereby reducing the pumping costs.

When the mixture of gas and said dilute aqueous solution reaches the rig, it is separated, and water vapour is boiled off from said dilute aqueous solution in order to regenerate said aqueous solution for the recycling and reuse.

5

Steam generated from regenerating said aqueous solution can be used for heating said aqueous solution for re-injection, in order to increase the rate of dissolution of the hydrate. Alternatively, the steam can be used for other 10 heating or power generation requirements.

In a preferred embodiment, the method includes the step of subjecting the hydrate formation to hydraulic fracturing with a fracturing fluid in order to depressurise the hydrate 15 formation and thereby facilitate the liberation of trapped hydrocarbon, wherein the fracturing fluid comprises an aqueous solution of a formate salt of any alkali metal or an acetate salt of any alkali metal, or a mixture of any two or more thereof. Preferably, said fracturing fluid has the same 20 composition as said aqueous solution.

Fracturing is a well known method for accessing subterranean formations and for stimulating the production of hydrocarbons from such formations. Fracturing fluid is 25 pumped from the surface and down the well drilled into the formation at a sufficiently high pressure to overcome the natural confining pressures in the formation and to induce a fracture in the formation. The opening created will remain open as long as sufficient fluid is injected at a high enough

pressure to overcome the effect of fluid seepage from the walls of the induced fracture.

In order to maintain the fracture open after the release
5 of injection pressure, a granular "proppant" with a high
resistance to crushing such as sand, bauxite or the like may
be added to the fracturing fluid. The proppant provides a
permanent support, propping up the fracture in the absence of
pressurised fracturing fluid. In some circumstances, it is
10 advantageous to continue to inject fluid under pressure into
the fracture, in which case a proppant is unnecessary.

The fracturing fluid may comprise a water soluble polymer such as a polysaccharide, in order to increase the
15 fluid viscosity. Suitable polysaccharides include guar gum, xanthum gum, or cellulose derivatives such as hydroxyethyl cellulose. Rheological properties can be further enhanced by cross-linking the polymers to increase the polymer viscosity by adding suitable cross-linking agents such as borate salts.
20 High viscosity increases the size of the induced fracture, helps limit fluid loss from the fracture through seepage, and helps carry the proppant. A pH buffer may also be added to enhance the action of the polysaccharide polymer and the cross linker.

25

An oxidising agent such as ammonium persulphate may be added to break the cross-linking and thereby lower the viscosity once the fracturing process is concluded, so that the fracturing fluid can be removed. Alternatively an enzyme

may be employed to break specific cross-linking bonds in the polymer.

The skilled addressee will be able to select suitable
5 additives for the fracturing fluid from the common general
knowledge in the art.

The advantage of using the claimed aqueous solutions as
fracturing fluids are that they remain in solution in the
10 induced fractures at very low temperatures, because they have
much lower freezing points than known fracturing fluids.

In a further aspect of the invention, there is provided
the use of a formulation comprising an aqueous solution of a
15 formate salt of any alkali metal or an acetate salt of any
alkali metal, or a mixture of any two or more thereof, as a
fracturing fluid for hydraulic fracturing. Preferably, the
formulation has the same composition as said aqueous solution
described above. More preferably, the formulation
20 additionally comprises a water soluble polymer such as a
polysaccharide, in order to increase the viscosity of the
formulation as described above.

In yet a further aspect of the invention, there is
25 provided a fracturing fluid for hydraulic fracturing in sub-
sea formations, comprising an aqueous solution of from 10% to
75% by weight of a formate salt of any alkali metal or an
acetate salt of any alkali metal, or a mixture of any two or
more thereof, and from 1 to 12 kg of a water-soluble polymer,
30 preferably a polysaccharide, most preferably guar gum, per

cubic metre of fracturing fluid. The fracturing fluid may also comprise a cross-linking agent for said polymer, in order to increase the viscosity of the fracturing fluid. Other additives for the fracturing fluid will be part of the 5 common general knowledge of the skilled addressee.

The method in accordance with the invention may be used with a single well or with a plurality of wells. Researchers have proposed using a two well system which has one injection 10 well and one production well connected by a horizontal fracture. This is only possible however with very shallow hydrate formations where induced horizontal fracturing is possible. As is described above, the horizontal fracture can be maintained at a very low temperature by using the salt 15 solutions described as fracturing fluids.

The decomposition of gas hydrate increases the level of gas production from underlying gas reservoirs. Depressurisation of the hydrates will enhance production as 20 decomposition progresses.

Preferred embodiments of the invention will now be described, with reference to the drawing, in which:-

25 Figure 2 shows a schematic diagram of the method in accordance with the invention.

Referring to Figure 2, a well bore 7 is drilled through impermeable cap rock 1 and permafrost 2 into hydrate 30 formation 3 overlying natural gas reservoir 4. Well bore 7

is lined with cement seal 8 to prevent the escape of gas from well bore 7.

In use, injection booster pump 15 injects an aqueous
5 solution of 65% by weight of potassium formate from the rig down injection line 16, through well bore 7 and into hydrate formation 3. The concentrated potassium formate solution acts to dissolve the hydrate formation 3, thereby creating a dissolution zone 5 comprising natural gas, water vapour, and 10 potassium formate solution. These components are pumped up tubing production string 6 by electrical submersible pump 18, and back to the rig along a pipeline.

The concentrated potassium formate solution absorbs
15 water vapour in the pipeline, thereby inhibiting the formation of gas hydrates.

The mixture of natural gas and dilute formate solution is separated by separator 9, and most of the gas is piped for 20 shipment or storage along pipeline 10.

The dilute formate solution is piped along pipeline 11 to boiler 12, which is powered by a proportion of the recovered gas. The dilute formate solution is boiled in 25 boiler 12 to regenerate concentrated formate solution and water. The water is stored in tanks 17, and the concentrated formate solution (which is now at an elevated temperature) is piped to a heat exchanger 14, where the excess heat is either used to preheat the injection line or sent by a steam along

steam line 13 for power generation and/or waste heat provision.

Prior to recycling, sufficient potassium hydroxide is
5 added to the formate solution to stabilise the pH of the
solution in the range pH 8-11, in order to reduce its
corrosive tendencies. Alternatively, a corrosive inhibitor
can be added. The concentrated formate solution can then be
sent back to the well bore for re-use.

CLAIMS

1. A method for recovering hydrocarbon trapped in a hydrate formation, comprising the steps of

5

(a) contacting the hydrate formation with an aqueous solution comprising from 10% to 75% by weight of a formate salt of any alkali metal or an acetate salt of any alkali metal, or a mixture of any two or more thereof, whereby said aqueous solution liberates hydrocarbon from the hydrate formation;

10

(b) transporting liberated hydrocarbon and at least part of said aqueous solution to a separator, wherein during the transportation step said aqueous solution absorbs at least part of any water vapour present to form dilute aqueous solution, thereby inhibiting formation of hydrocarbon hydrates;

15
20

(c) separating hydrocarbon from said dilute aqueous solution;

(d) regenerating said aqueous solution by heating said dilute aqueous solution to remove absorbed water vapour; and

25

(e) recycling said aqueous solution to step (a).

2. A method as claimed in Claim 1, including the step of subjecting the hydrate formation to hydraulic fracturing with a fracturing fluid in order to depressurise the hydrate

formation and thereby facilitate the liberation of trapped hydrocarbon, wherein the fracturing fluid comprises an aqueous solution of from 10% to 75% by weight of a formate salt of any alkali metal or an acetate salt of any alkali metal, or a mixture of any two or more thereof.

3. A method as claimed in Claim 2, wherein said fracturing fluid has the same composition as said aqueous solution of Claim 1.

10

4. A method as claimed in any one of the preceding claims, wherein said aqueous solution comprises at least 40% by weight of said salt.

15

5. A method as claimed in any one of the preceding claims, wherein said aqueous solution comprises from 40% to 65% by weight of said salt.

20

6. A method as claimed in any one of the preceding claims, wherein said alkali metal is potassium, sodium, rubidium or caesium.

7. A method as claimed in any one of the preceding claims, wherein said salt is potassium formate.

25

8. A method as claimed in any one of the preceding claims, wherein said aqueous solution is heated prior to contact with the hydrate formation.

9. A method as claimed in any one of the preceding claims, wherein said aqueous solution is injected into the hydrate formation through an insulating jacket, the jacket having a layer of insulation fluid comprising an aqueous solution of from 10% to 75% by weight of a formate salt of any alkali metal or an acetate salt of any alkali metal, or a mixture of any two or more thereof.

10. A fracturing fluid for hydraulic fracturing in sub-sea formations, comprising an aqueous solution of from 10% to 75% by weight of a formate salt of any alkali metal or an acetate salt of any alkali metal, or a mixture of any two or more thereof, and from 1 to 12 kg of a water-soluble polymer per cubic metre of fracturing fluid.

15 11. A fracturing fluid as claimed in Claim 10, wherein the water soluble polymer is a polysaccharide.

12. A fracturing fluid as claimed in Claim 10 or Claim 11,
20 wherein the water soluble polymer is guar gum.

13. A fracturing fluid as claimed in any one of Claims 10 to 12, additionally comprising a cross-linking agent for said polymer.

25 14. The use of a formulation comprising an aqueous solution of from 10% to 75% by weight of a formate salt of any alkali metal or an acetate salt of any alkali metal, or a mixture of any two or more thereof as a fracturing fluid for hydraulic
30 fracturing.

15. The use of an aqueous solution of from 10% to 75% by weight of a formate salt of any alkali metal or an acetate salt of any alkali metal, or a mixture of any two or more thereof, as a dehydrating agent for a hydrocarbon.

5

16. A method for recovering hydrocarbon trapped in a hydrate formation substantially as hereinbefore described, with reference to the drawings..



FIGURE 1

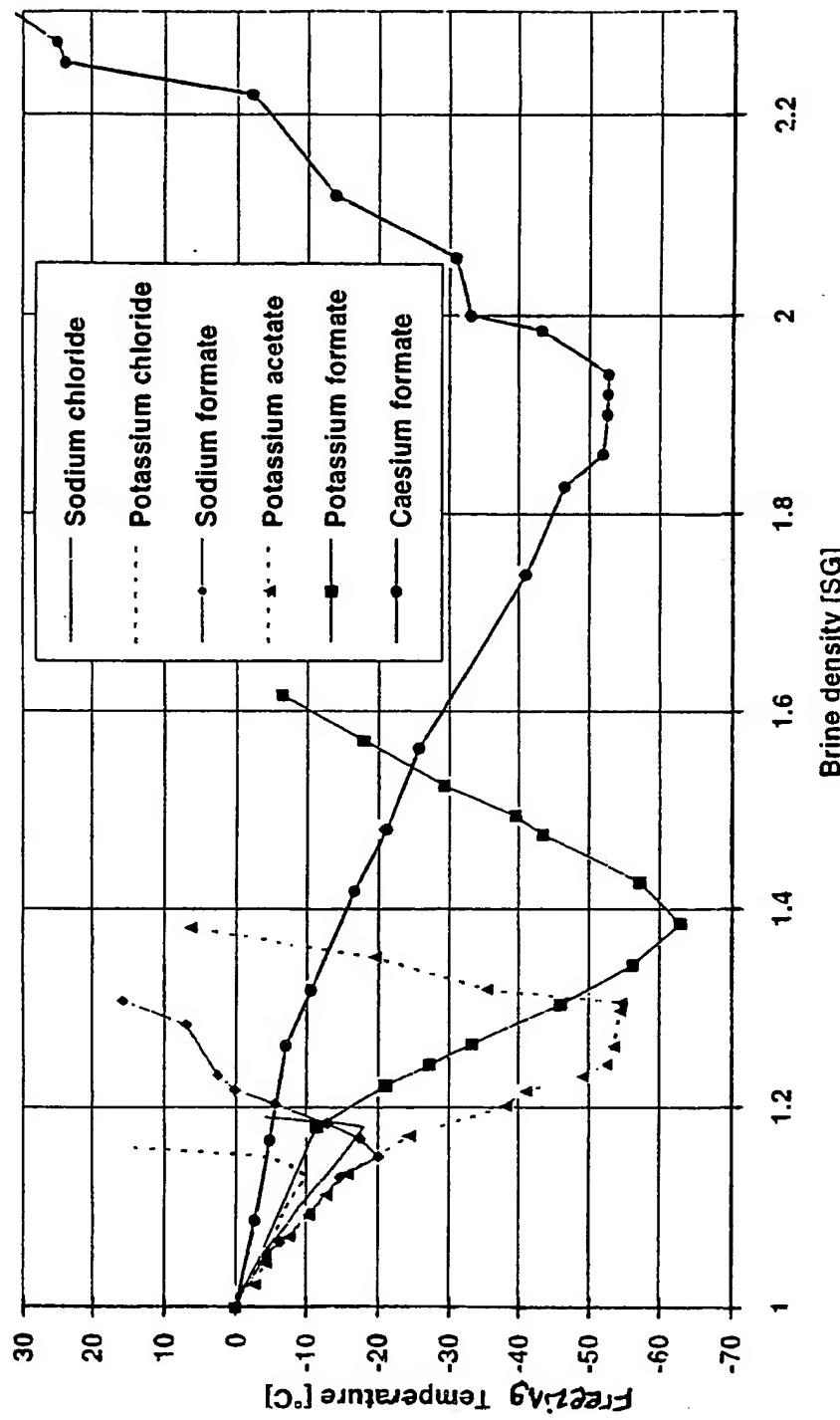
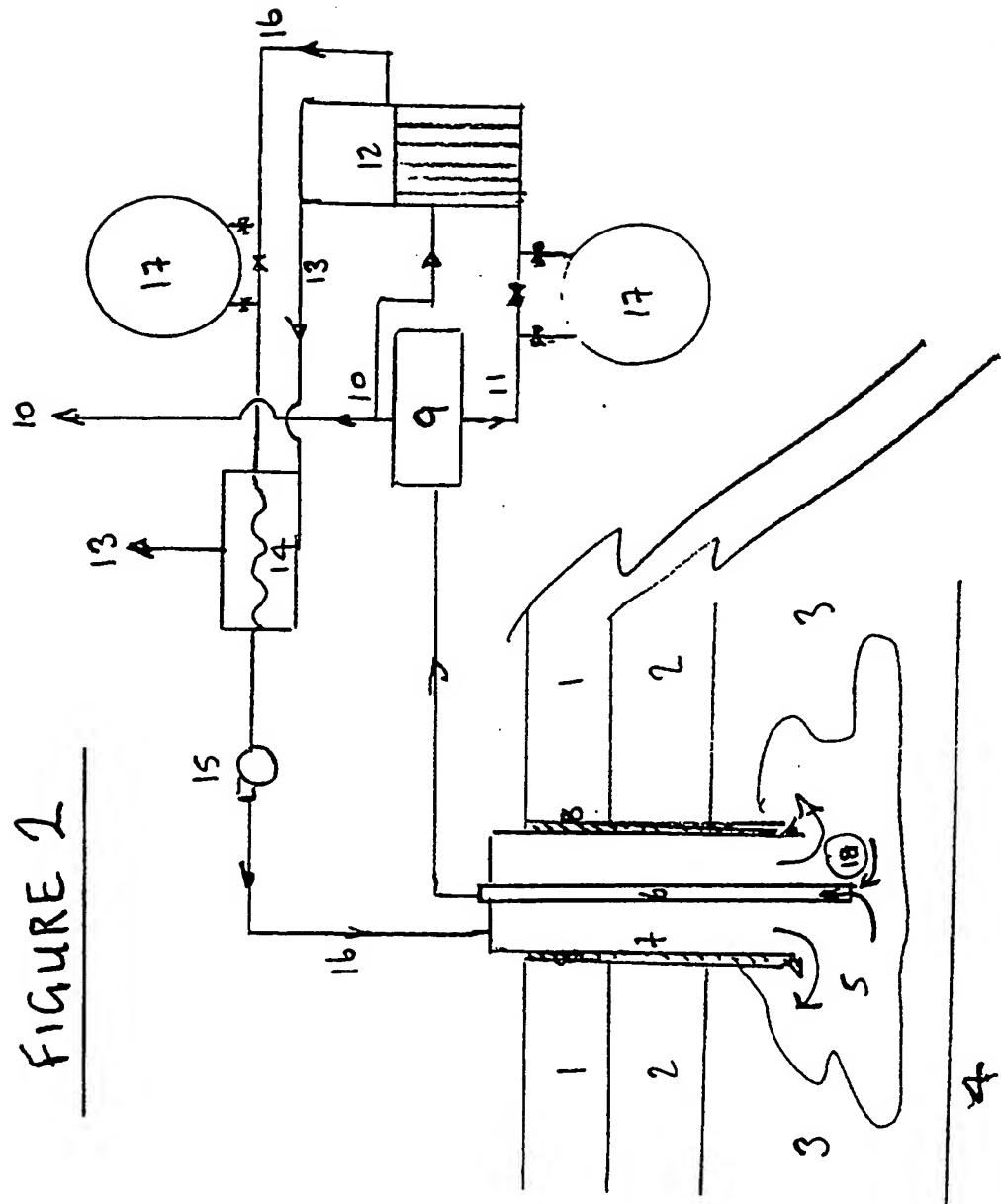


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